THE JOURNAL OF PREVENTIVE MEDICINE
2001; 9(3): 3-14

NATURAL RADIOACTIVITY OF SOME MINERAL WATERS AND POPULATION DOSES

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Abstract. There are many mineral water springs in the northeastern Romania which, traditionally, are used as table mineral water sources in the area. This study has as basic objectives the radioactivity control of the drinking mineral waters according to existing standards and evaluation of doses to population by ingestion of mineral water. During the period from 1997 to 2000, 107 water samples (bottled waters commercially available for human intake and some spring waters) were collected. Their total alpha and beta radioactivity and the natural radioelements (natural uranium and thorium, radium-226 and potassium-40) were measured. The following contents were found: 0.25 – 35 mBq/l for ^{238}U; 0.04 – 4.4 mBq/l for ^{232}Th and 9 – 1250 mBq/l for ^{40}K. The corresponding activities of ^{226}Ra up to 1 Bq/l and concentrations of gross alpha radioactivity up to 3 Bq/l were also measured. The values obtained were compared with the reference ones accepted for drinking water. A calculation of dose based on radioactive content data and the ICRP (International Commission on Radiological Protection) dozimetric model for ingestion was performed. The effective dose values due to the intake of these radioelements through water consumption were estimated to range from about 1.5 to 28 μSv/y. The resulting individual annual effective doses are very low quite insignificantly versus other natural sources. The contribution of mineral water to overall exposure from natural sources is only of 0.25 % up to 1.2 %.

Key words: mineral water, springs, bottled water, natural radioactivity, gross alpha and beta radioactivity, specific activities of natural uranium, natural thorium, radium-226, potassium-40, effective doses

Rezumat. Există numeroase izvoare de apă minerală în Nord-Est-ul României, care sunt utilizate ca ape de masă. Scopul acestui studiu a fost de a determina conţinutul radioactiv al acestor ape şi contribuţia ingestiei lor la expunerea populaţiei. Prelevarea şi analiza probelor s-a efectuat conform standardelor în vigoare. Am prelevat probe de apă provenind din izvoare care sunt utilizate de localnici ca apa potabilă carbogazoasă şi probe de apă care se comercializează în butelii de sticlă sau plastic, şi sunt plate sau carbogazoase (natural sau prin adăugare de CO_2). Prelevarea probelor de apă minerală s-a efectuat direct de la sursă iar în cazul probelor de apă de masă care se comercializează, s-au recoltat în paralel probe de apă minerală îmbuteliată. După determinarea radioactivităţii alfa şi beta globale, toate probele au fost analizate din punct de vedere al conţinutului de ^{226}Ra, uraniu natural (respectiv uraniu-238), toriu natural (respectiv toriu-232) şi potasiu-40. Domeniile de variaţie ale concentraţiilor de activitate găsite în probele analizate au fost următoarele:

- Activitate globală alfa: (8–3.100) (mBq/l), activitate globală beta: (22–2.470) mBq/l
- Uraniu natural: (0,51 – 75) mBq/l, Uraniu-238: (0,25 – 35) mBq/l
- Radiu-226: (1,2 – 1.035) mBq/l
- Toriu natural: (0,05 – 5,3) mBq/l, Toriu-232: (0,04 – 4,4) mBq/l

- Potasiu-40: (9 – 1250) mBq/l

Rezultatele obţinute pentru probele de apă minerală se situează în domeniile de maxim ale valorilor concentraţiilor de activitate ale apei potabile din surse de profunzime (fântâni sau microcentrale) din Moldova. În niciuna din probe nu s-a depăşit activitatea specifică admisă în apa potabilă pentru uraniu, toriu natural şi potasiu-40. Concentraţia de radiu-226 a atins în câteva probe activitatea admisă excepţional (500mBq/l), ajungând în două probe până la 1000 mBq/l. Aceasta valoare, conform CIPR (Comisia Internaţională de Protecţie Radiologică), este o concentraţie derivată pentru un grup de indivizi şi reprezintă nivelul maxim permisibil pentru ^{226}Ra în apa potabilă. Dozele efective individuale de iradiere s-au calculat luând în considerare conţinutul de radioelemente naturale, un consum de 0,5 L/zi şi coeficienţii de conversie specifici ingestiei şi fiecărui radioelement conform CIPR. O persoană adultă din populaţie care consumă 0,5 L/zi apă minerală de masă primeşte în plus 6 μSv/an faţă de fondul natural de iradiere de 2450 μSv/an din toate sursele naturale de iradiere pentru Moldova (Nord-Estul României). Contribuţia apei minerale la iradierea naturală a populaţiei reprezintă 0,25 % până la 1,2 % din fondul natural de iradiere.

Cuvinte cheie: apă minerală, izvoare, apă îmbuteliată, radioactivitate alfa şi beta globală, radioactivitate naturală, activitate specifică a uraniului şi toriului natural, ^{226}Ra, ^{40}K, doze efective

INTRODUCTION

As a rule, mineral water springs run across highly mineralized rocks. The geological sources of natural mineral water are known as aquifers, which may be of different types, and they vary greatly in terms of their depth, horizontal extent, composition, and permeability. Water filtering into the underground flows slowly through deep permeable rocks and sediments and diffuses into the empty interstitial space of the rocks. While passing through the underground strata, water picks up minerals and other elements depending on the chemical make-up of the strata. That is why they have higher concentrations of minerals, trace minerals and natural radioelements than other kinds of water.

The most radionuclides are minerals dissolved in water (Dissolved Mineral Radioactivity). Radioactive minerals occur irregularly in the bedrock, similar to other minerals and they dissolve easily in water. Bedrock contains naturally occurring radioactivity including uranium, thorium, radium and potassium. The natural radioactivity results from water passing through deposits of naturally occurring radioactive materials. In some areas this causes the groundwater and underground water to exceed current or proposed public drinking water standards for radioactivity.

Natural mineral water differs, in its original purity and its content from treated water what we drink. The surface and below ground fresh water sources undergo a treatment process (settlement, filtration, precipitation, purification etc.). During these appropriate treatments for meeting specific bacteriological and chemical safety standards, the possible radionuclides are retained. As contrasted with this situation the mineral waters come to us in an unadulterated way. Bottling is done at the source and treatments to modify the composition of or purify natural mineral water are prohibited (1-4).

Numerous data published in literature describe water as the main vector of transfer for natural radioelements from environment to human beings. As mineral waters are widely used as drinking water we were interested in finding out the extent to which these waters can be a natural radiation source.

Radioactive minerals exist in certain areas in northeastern Romania. There are many mineral water springs which, traditionally, are used as mineral water sources in the area. This survey aims to assess the radioactive content of these waters and their contribution to the population exposure.

MATERIAL AND METHODS

The mineral water springs clustered in northeastern Romania as well as their uses have been inventoried. During the period from 1997-2000 water samples (bottled waters commercially available in two styles, still and sparkling, and some spring waters) in Neamt, Iasi and Suceava districts were collected several times over this period at all locations of mineral water. In all samples total alpha and beta radioactivity and the natural radioelements of utmost interest (natural uranium and thorium, radium-226 and potassium-40) were measured.

The sampling and samples analysis were carried out using standardized methods, in Romania (3-10). The mineral water samples were directly obtained from spring or from bottling station, collected in polyethylene bottles, and were analyzed without any addition or previous treatment.

The water samples were concentrated by means of active coal absorption and evaporated. The gross alpha particle activities of water sample were measured by using a ZnS (Ag) detector system and a plastic scintillation for the gross beta activities, respectively. Efficient techniques of element specific separation were developed allowing the analysis of each element. Uranium-238 and thorium-232 levels were calculated after determination of content in natural uranium and thorium using a method based on their separation and purification on a strong basic anion exchange resin and spectrophotometric measurement in terms of the Arsenazo III complex. Radium-226 was determined through its decay product radon-222, and by alpha ray measurement in a scintillation chamber. Levels of potassium-40 were determined by means of calculation following photometric dosing in the flame emission mixture of natural potassium isotopes.

The total dissolved solids (TDS) are usually measured as the residues (what remains) when a liter of water is evaporated at 180^0 C. The TDS were determined in all samples by evaporation to dryness and weighting the residue.

Based on the mean concentrations values of natural radionuclides in the mineral water and using the conservative approach of a single intake the effective doses were estimated. Doses resulting from the consumption of these waters were calculated using ICRP dose conversion coefficients (11-13).

RESULTS AND DISCUSSION

Mineral waters contain - as suggest the name - various minerals and trace elements. Some of analyzed mineral waters were with "low mineral content" and others with "high mineral content". The TDS content of mineral water ranged from 40 to 2,700 mg/l. The carbonated samples tended to show higher TDS values.

The analysis of the got radioactivity data shows that natural radioactivity in table mineral water varies over a large range (up two orders of magnitude). An explanation for the different concentrations of natural radioelements in bottled mineral water and spring water relating to temperature, dissolved inorganic salts, geological composition and other factors can be suggested.

Some values exceed the Romanian water quality guidelines for drinking water regarding gross alpha activity and radium-226 (7). The natural mineral water has been included in the existing Romanian legislation on drinking water and radioactivity control is compulsory (4).

Table 1 shows the minimum and the maximum levels of each radiological indicator for drinking mineral water, comparatively with maximum admitted concentrations (MAC) and maximum admitted specific activities (MASA) used in Romania on drinking water (7).

Table 1. The activity concentrations and specific activities (mBq/l)

Radioactive elements	Table mineral water (range)	Maximum Concentrations		Maximum Specific activity	
		Admitted	Exceptionally Admitted	Admitted	Exceptionally Admitted
Gross Alpha radioactivity	8 – 3,100	100	2,300		
Gross Beta radioactivity	22 – 2,470	800	50,000		
Natural Uranium	0.51 – 75			590	1,000
Uranium-238	0.25 – 35				
Radium-226	1.2 – 1,035			88	500
Natural Thorium	0.05 – 5.3			40	100
Thorium-232	0.04 – 4.4				
Potassium-40	9 – 1,250			13,420	-

The values of measured concentrations are comparable but lower than concentration values reported for other countries (14-33).

The values of the Gross Alpha and Beta radioactivity in most of the water samples are within the drinking water standard, even if exceptionally admitted (7). However, of all samples collected, 83 % have an alpha-activity higher than MAC (100 mBq/l) and only 15 % have a beta-activity higher than MAC (800 mBq/l). No relation between the ^{226}Ra concentrations and gross-alpha activity were obtained. On the other hand, the higher ^{40}K concentrations, the higher gross-beta activity was almost permanently found.

One should remark that neither of the analyzed samples exceeds the specific activity admitted in Romania for fresh water, in any of the natural radioelements under investigation excepting ^{226}Ra. Some measured radium-226 concentrations exceed the reference values of 88 mBq/l accepted for drinking water and the exceptionally admitted ones of 500 mBq/l, even.

The highest values of uranium and radium were obtained especially in springs originating from a depth higher 60 meters and lead one to conclude that radioactive content is mainly related to the mineralization in waters of underground origin. Waters that presented high levels of carbonate and sulfate salts showed maximum values for ^{226}Ra. This behaviour is mainly due to the physico-chemical properties of this radionuclide in water as well as to the lithologic structure of the aquifers. (26).

The highest levels of ^{226}Ra and natural uranium concentration correspond to those found at the most in drinking ground waters from drilled wells in Neamt and Suceava districts the values being in good agreement with our data published previously (34,35). From among the ground water sources, the wells situated in the northeastern part of Romania, where the Neamt and Suceava districts are located, generally showed the highest concentrations in natural radioelements, especially with regard to uranium (up to 37 mBq/l) and radium-226 (up to 81 mBq/l). This may be explained by the fact that the ground and the rocks, on which the soil was formed in that area, are richer in natural radioelements than is the case for most part of Romania.

In Tables 2-4 are shown the average values and standard deviation of concentrations, calculated for natural radioelements that were measured in the drinking mineral water samples analyzed:

- The caught springs, sources for bottled table mineral water (*)
- The marketed table water - packed in the polymeric bottles (▲)
- Some communal springs (◊). The communal springs studied are open to public and frequently used as drinking water without any sanitary certificate.

Comparing all the figures, it can be seen a similar situation that is the low concentration of ^{232}Th. In most cases, the comparatively higher values for uranium and radium concentrations were observed in the water samples from Suceava district. Only two springs, located in Suceava district, sources for Bucovina brand (Tables 1 and 3), exceeded the allowed maximum level for an individual group of 1000 mBq/l for ^{226}Ra (12). This can be explained by fact that mineral waters of the Na-Ca-Cl type, like Bucovina's source, provide favorable conditions for the mobilization of radium.

Table 2. Natural radioactivity (m ± SD) in natural mineral waters in Iaşi and Neamț district (in mBq/l)

Water sample (source* and brand▲)	Natural Uranium	^{238}U	^{226}Ra	^{232}Th	Gross Alpha-radioactivity	^{40}K	Gross Beta-radioactivity
Iaşi district							
Spring 3 in Botanical Gardens*	27.6 ± 15.1	13.1 ± 7.3	12.6 ± 2.8	1.2 ± 0.4	139 ± 46	170 ± 31	308 ± 65
Amfiteatru (still)▲	28.3 ± 13.9	13.6 ± 6.6	15.7 ± 6.1	0.57 ± 0.004	81 ± 14	190 ± 22	176 ± 84
Amfiteatru (sparkling)▲	22.8 ± 4.7	11.2 ± 2.3	14.2 ± 8.9	1.4 ± 1.1	95 ± 46	150 ± 30	176 ± 65
Neamț district							
Toşorog-Bicaz*	27.0 ± 12.0	13.0 ± 6.0	15 ± 6.8	2.5 ± 0.8	74 ± 9	260 ± 52	360 ± 84
Carpatina forte▲	12.4 ± 2.1	6.2 ± 1.1	9.0 ± 0.2	1.68 ± 0.22	42 ± 11	160 ± 42	320 ± 74

Table 3. Natural radioactivity (m ± SD) in natural mineral waters in Suceava district (in mBq/l)

Water sample (source* and brand▲)	Natural Uranium	^{238}U	^{226}Ra	^{232}Th	Gross Alpha-radioactivity	^{40}K	Gross Beta-radioactivity
Poiana Vinului*	29.3 ± 19.5	14.3 ± 5.7	23.3 ± 4.8	0.92 ± 0.26	284 ± 102	112 ± 7	108 ± 47
Dorna▲	13.2 ± 5.1	6.2 ± 2.3	3.4 ± 1.7	0.061± 0.014	111 ± 28	91 ± 40	< 100
Poiana Negri*	18.6 ± 4.1	9.0 ± 1.8	1.5 ± 0.7	0.082 ± 0.031	283 ± 72	737 ± 101	304 ± 135
Poiana Negri▲	18.1 ± 7.8	8.7 ± 3.2	1.1 ± 0.18	0.17 ± 0.028	127 ± 84	790 ± 120	675 ± 120
Dorna Candreni*	8.3 ± 4.3	4.0 ± 2.1	2.05 ± 0.41	0.35 ± 0.16	1856 ± 432	59 ± 20	638 ± 116
Cristalina▲	16.6 ± 1.0	8.0 ± 0.7	7.72 ± 4.61	1.23 ± 0.56	1150 ± 320	46 ± 10	1600 ± 437
Cheson C_3, C_7 Devil's mill*	18.0 ± 5.9	8.7 ± 1.2	9.5 ± 4.4 90.0 ± 20.0	0.38 ± 0.09	8 – 520[a]	27 ± 10	72 ± 14 87 – 2470[a]
F_1, F_2 Red spring, Secu*	20.4 ± 8.1	7.8 ± 1.9	23.2 ± 18.1 **576 ± 394**	0.93 ± 0.41	12 – 3140[a]	136 ± 71	22 – 760[a]
Bucovina (still) ▲	18.2 ± 4.4	8.8 ± 2.1	65.0 ± 7.0 154.0 ± 64.0	0.83 ± 0.26	12 – 492[a]	51 ± 21	57 – 470[a]
Bucovina (carbonated)▲	11.5 ± 5.6	5.5 ± 2.4	23.05 ± 10.1 82.0 ± 19.0	0.78 ± 0.49	8 – 181[a]	94 ± 52	60 ± 18

a = range

Table 4. Natural radioactivity (m ± SD) in communal springs (◊) of mineral water (in mBq/l)

Sample location	Natural Uranium	^{238}U	^{226}Ra	^{232}Th	Gross alpha radioactivity	^{40}K	Gross beta radioactivity
Iasi district							
2 and 5 springs in Botanical Gardens Iasi	25.1 ± 13.2	12.1 ± 6.4	11.4 ± 2.6	0.61 ± 0.09	160 ± 38	214 ± 64	244 ± 39
Pârcovaci Springs	6.6 ± 1.9	3.2 ± 0.9	6.1 ± 3.8	0.29 ± 0.01	118 ± 96	330 ± 31	240 ± 40
Neamt district							
Borca	16.5 ± 1.6	8.0 ± 0.7	17.5 ± 0.4	0.91 ± 0.09	89 ± 23	49 ± 16	45 ± 12
Almas Garcina	30.9 ± 15.5	14.9 ± 8.2	1.1 ± 0.5	0.68 ± 0.03	38 ± 14	21 ± 6	63 ± 14
Grinties springs	22.8 ± 21.4	11.0 ± 10.1	27.2 ± 16.4	0.41 ± 0.03	173 ± 58	115 ± 34	126 ± 40
Suceava district							
S spring Vatra Dornei	14.3 ± 7.3	6.0 ± 3.8	4.8 ± 1.7	0.57 ± 0.21	25 ± 15	51 ± 9,2	87 ± 16
B spring Vatra Dornei	39.1 ± 5.7	18.9 ± 7.3	0.32 ± 0.20	3.35 ± 0.32	79 ± 17	17 ± 8	57 ± 19
Neagra Broșteni	9.5 ± 2.6	9.4 ± 2.1	40.4 ± 19.7	< 0.082	105 ± 34	162 ± 56	495 ± 231
Crucea	51.0 ± 12.6	24.5 ± 6.1	72.0 ± 18.0	0.57 ± 0.21	72 ± 11	84 ± 23	84 ± 14
Smalțu-Dârmocsa	< 1.014	< 0.49	4.2 ± 2.4	< 0.082	32 ± 8	459 ± 79	838 ± 167

Table 5. Average activity concentrations (m ± SD) of natural radionuclides in drinking mineral water samples (in mBq/l) (County Moldavia – Romania 1997 - 2000)

District	U_{nat}	^{238}U	^{226}Ra	^{232}Th	^{40}K	Notes
SUCEAVA	24.8 ± 8.5	12.0 ± 4,1	19.0 ± 16.0	0.61 ± 0.52	220 ± 175	All brands
	16.1 ± 3.7	8.2± 2.0	226.0±149.0	0.66 ± 0.27	90 ± 51	Bucovina brand
	27.1 ± 17.0	13.2 ± 8.2	24.3 ± 26.1	0.86 ± 0.74	120 ± 87	Communal springs
IAŞI	25.6 ± 8.4	12.4 ± 4.1	11.5 ± 6.6	1.25 ± 0.48	178 ± 56	Amfiteatru brand
	20.3 ± 10.4	9.8 ± 2.4	8.7 ± 4.4	0.49 ± 0.12	272 ± 81	Communal springs
NEAMŢ	19.7 ± 9.1	9.6 ± 4.9	12.1 ± 3.2	2.07 ± 0.40	210 ± 70	Carpatina brand
	23.4 ± 7.2	11.3 ± 3.1	15.3 ± 10.2	0.67 ± 0.25	78 ± 42	Communal springs

Table 6. Individual average effective doses (m ± SD) due to the consumption of mineral water (in μSv/y)

District	U_{nat}	^{226}Ra	^{232}Th	^{40}K	Total	Notes
SUCEAVA	0.196 ± 0.066	1.13 ± 0.95	0.13 ± 0.11	0.25 ± 0.19	1.71 ± 0.99	All brands
	0.135 ± 0.033	13.4 ± 8.9	0.14 ± 0.06	0.10 ± 0.06	13.8 ± 8.9	Bucovina brand
	0.217 ± 0.135	1.44 ± 1.55	0.19 ± 0.16	0.13 ± 0.10	1.98 ± 1.57	Communal springs
IAŞI	0.202 ± 0.066	0.68 ± 0.39	0.27 ± 0.10	0.20 ± 0.06	1.35 ± 0.41	Amfiteatru brand
	0.161 ± 0.039	0.52 ± 0.26	0.11 ± 0.03	0.31 ± 0.09	1.10 ± 0.28	Communal springs
NEAMT	0.157 ± 0.081	0.72 ± 0.19	0.45 ± 0.09	0.23 ± 0.08	1.56 ± 0.23	Carpatina brand
	0.75 ± 0.23	3.68 ± 2.46	0.59 ± 0.22	3.76 ± 2.01	8.78 ± 1.25	Communal springs

Such waters are expected to contain generally enhanced radium concentrations even in regions with moderate levels of natural radioactivity (29).

The values of geometric means for ^{226}Ra are of 32.6, 11.6 and 8.2 mBq/l in Suceava, Neamt and Iasi district, respectively.The geometric means for uranium-238 have values of 8.7, 6.7 and 6.3 mBq/l , respectively for the same districts.

The values are within the range of average world values (14,15). The radium-226 concentrations in bottled mineral water from European countries range up to 5,500 mBq/l and have geometric means of 7, 14, 27, 44 mBq/l in Italy, Sweden, Portugal, France, respectively and 25 mBq/l in Austria and Germany. Uranium-238 concentrations in European mineral water are similar to those observed in USA's ground water, with median value of 12 mBq/l for France and geometric means of 4 up to 24 mBq/l for Germany. In Slovenia and Croatia were reported radium-226 values ranging from 5 to 510 mBq/l for underground and mineral water. (19,24) The following contents of 30-720 mBq/l for ^{234}U+^{238}U, < 1-5 mBq/l for ^{232}Th and 5-370 mBq/l for ^{226}Ra were determined in Switzerland. (21) In Argentina and Brazil, the reported values of the natural radioelements concentrations in mineral water are higher than those founded for European countries (22,23,26).

Internal exposure arising from the intake of long-lived natural radionuclides through the ingestion of drink mineral water was evaluated taking into account the natural radioactive content of water and the yearly average consumption of 1,825 liters (around 0.5 l/day). The dose conversion coefficients endorsed by ICRP (12,13) for each radioelement were applied.

As the large variability of activity concentrations in samples, with a view to estimate the radiation doses, the mean values were calculated (see Table 5).

The average values of individual effective dose rates through ingestion of mineral water are presented in Table 6. These estimates are of the order of minim 1.5 μSv y^{-1} and maxim of 30 μSv y^{-1}, depending on the geographical region.

CONCLUSIONS

- An individual which drinks mineral water 0.5 l/day gets the average of 6 μSv·y^{-1} over background radiation of 2,450 μSv·y^{-1} due to all natural radiation sources in Moldova (northeastern of Romania). Ingestion of mineral water contributes to natural radiation exposure with 0.25 % up to maxim 1.2 % as part of background radiation.
- Notwithstanding this, in order to obtain license concerning the commercialization of mineral waters as drinking water, an evaluation of the radioactive levels, consisting in determination of radium-226 concentration and global alpha and beta activity should be performed.

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